PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No.

10/716,894

Confirmation No. 9412

Applicant(s)

Cornelis F. VAN EGMOND et al.

Filed

: November 19, 2003

.....

1401011111111

TC/A.U.

; 1764

Examiner

: In Suk C. BULLOCK

Title

Methanol and ethanol production for an oxygenate to olefin reaction system

Arry. Docket No. :

2003B113

Customer No.

23455

Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

DECLARATION SUBMITTED UNDER 37 C.F.R. § 1,132

Dear Sir:

I, Teng Xu, a U.S. citizen, residing at 13422 Scenic Glade Drive, Houston, TX 77059, declare and state that:

- 1. I received a Ph.D. in Chemistry at the University of Texas A&M University.
- Since 1998, I have engaged in scientific research at ExxonMobil Chemical
 Company in the field of Methanol Conversion to Olefins on Molecular Sieve Catalysts.
- I am author or co-author of several publications dealing with converting oxygenates such as methanol to olefins on zeolite solid acids.
- 4. I am one of the joint inventors of U.S. Patent Application No. 10/716,894 (the '894 application) and am familiar with the subject matter thereof.

BEST AVAILABLE COPY

NOV-21-2006 TUE 02:08 PM EXXONMOBIL NOV-21-2006 TUE 10:24 AM EXXONMOBIL

FAX NO. 281 834 7413 FAX NO. 281 834 2073

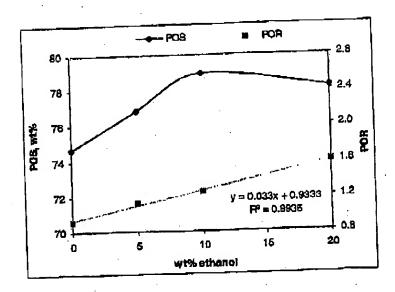
P. 16 P. **0**2

Appl. No. 10/716,894 Atty, Docket No.: 2003B113 Office Action dated August 1, 2006 1.132 Decl. w/Suppl. Amdmt. dated Nov. 21, 2006

- 5. I have read U.S. Patent No. 6,441,262 (Fung) cited in the July 17, 2006 Office Action and am familiar with the subject matter thereof.
- 6. The Fung reference discloses using a silicoaluminophosphate molecular sieve catalyst to convert a combination of methanol and ethanol to light olefins. The reference is particularly concerned with the problem of how to moderate the olefin product content in an alcohol conversion reaction in which the alcohol feed is predominantly methanol. Fung addresses this problem by using two contact zones. One zone is referred to as an oxygenate conversion zone, which is where a combination of unregenerated and regenerated catalyst contacts methanol for conversion to olefin. The other zone is referred to as an alcohol contact zone, which is where ethanol, propanol or butanol is contacted with regenerated and fresh catalyst. The use of this dual contacting zone allows for manipulating the olefin content in the product stream.
- 7. This invention is particularly concerned with maximizing the amount of light or prime clefin formed (i.e., ethylene plus propylene), and at the same time further maximizing the amount of ethylene in the product. This problem is solved by providing a mix of methanol and ethanol, at a 4:1 to 19:1 weight ratio of methanol to ethanol, to a reaction zone, and contacting that mix with a silicoaluminophosphate molecular sieve catalyst at a temperature of at least 475°C to 500°C, and possibly higher, in the same reaction zone.
- 8. Aside from the data in Examples II and III (especially Tables III and IV) of the '894 application, additional experiments were performed under my authority and control over the claimed range (4:1 to 19:1 weight ratio of methanol to athanol, particularly at 5 wt% ethanol to augment the data already described for 10 wt% and 20 wt% ethanol), following the procedures of Examples II and III. The experiments were performed at 475°C, 100 WHSV, and 25 psig (total pressure of mixed alcohol feed), the same density for methanol and ethanol being assumed in calculating WHSV. The results are shown in the following graph:

Page 2 of 4

Appl. No. 10/716,894 Any, Docket No.: 2003B113 Office Action dated August 1, 2006 1.132 Decl. w/Suppl. Amdmt. dated Nov. 21, 2006



- 10. From the data in the Examples of the '894 application and the data shown in the above graph, I conclude that:
 - a) the amount of ethanol in methanol feed has a significant impact on POR
 (prime olefin ratio, i.e., ethylene to propylene ratio);
 - b) there is almost a linear relationship between wt% ethanol in feed vs. POR;
 - e) the amount of ethanol in methanol feed has a minor impact on POS (prime olefin selectivity, i.e., percent of ethylens + propylene in product); and
 - d) POS is shows a peak beginning at about 5 wt% ethanol and extending at least up to about 20 wt% ethanol.
- 11. I further conclude from the dara that at a temperature of at least 475°C to 500°C, a mix of from 5 wi% to 20 wi% ethanol provides superior POS and POR, and that such results would not have been expected by reading the Fung reference.
- 12. It is also my opinion that the coupling of temperature with the particular mix of ethanol and methanol as a feed stream would to provide superior POS and POR is not mere optimization of a reaction process, because there are enough significant variables and the

Appl. No. 10/716,894 Atry. Docket No.: 2003B113 Office Action dated August 1, 2006 1.132 Decl. w/Suppl. Amdur. dated Nov. 21, 2006

complexity of the reaction process is so high that one would not have been able to easily reach such a finding.

13. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that theses statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the captioned application or any patent issued therefrom.

Further declarant sayoth not.

11 1 20 1200 6

Teng Xu

Page 4 of 4